

# EXPERIMENTAL DETERMINATION OF FORCED CONVECTIVE HEAT TRANSFER COEFFICIENT WITH NANOFLUID IN A PLAIN TUBE

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## ABSTRACT

Nowadays, lots of industrial and consumer product have heat transfer as their most vital process. But there is a limit of the heat transfer due to the inherently poor thermal performance of conventional fluids and restricted in developing energy efficient heat transfer fluids. With the strong need from industry force the need to build up superior heat transfer fluids with considerably higher thermal performance than are presently available. Hence, lots of research has been made by the scientists and engineers to break this fundamental limit by dispersing metallic and non-metallic nanoparticles in liquids. There are two ways to prepare nanofluid which is two step method and dilution process. In this study, the process that used to prepare the nanofluids is dilution process.  $\text{TiO}_2$  water based nanofluid was prepared at four different concentrations 1%, 1.5%, 2% and 3%. The experiment was conducted for the estimation of heat transfer coefficient (htc) of the nanofluids at constant heat flux and various Reynolds number from 10,000 to 30,000. The experiment was started with distilled water for validation of experiment set up. Additions of the nanoparticles to the water increase their heat transfer coefficient with maximum enhancement of 13.6% compared with pure water at 1.5% volume percentage. Experimental result were compared with previous result in literature study and found consistent with considerable deviation observed.

## ABSTRAK

Pada masa kini, banyak produk industri dan pengguna mempunyai pemindahan haba sebagai proses yang paling penting mereka. Tetapi terdapat had pemindahan haba kerana cecair konvensional mempunyai kadar pengaliran haba yang rendah untuk mencipta cecair untuk pemindahan haba yang berkesan. Permintaan tinggi daripada industri mendorong kepada penciptaan cecair yang mempunyai muatan haba tentu yang lebih tinggi daripada yang wujud sekarang. Jadi, banyak kajian telah dibuat oleh para saintis dan jurutera untuk melangkaui had asas cecair dengan mencampurkannya bahan logam dan bukan logam yang bersaiz nano. Terdapat dua cara untuk menyediakan cecair nano (nanofluids) iaitu penyediaan dua langkah dan proses pencairan. Cara yang digunakan untuk kajian ini adalah proses pencairan.  $\text{TiO}_2$  cecair nano berasaskan air disediakan dengan kepekatan isipadu sebanyak 1%, 1.5%, 2% dan 3%. Eksperimen ini adalah untuk mengkaji perkali pemindahan haba pada nombor Reynold dari 10,000 hingga 30,000. Eksperimen dimulakan dengan air untuk mengesahkan eksperimen yang telah disediakan. Penambahan partikel bersaiz nano didalam air telah menaikkan perkali pemindahan haba sehingga 13.6% berbanding dengan air tulen pada kepekatan 1.5%. Keputusan eksperimen ini telah dibandingkan dengan keputusan penyelidikan sebelum ini dan mendapati ianya konsisten.

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## LIST OF SYMBOLS

$D_i$	Inner diameter of the tube, ( $m$ )
$h$	Convective heat transfer coefficient, ( $W/m^2.K$ )
$k$	Thermal conductivity, ( $W/m.K$ )
$\mu$	Dynamic viscosity of the fluid, ( $kg/m.s$ )
$\rho$	Density of the fluid, ( $kg/m^3$ )
$C_p$	Specific heat, ( $J/kg.K$ )
$L$	Length of the tube, ( $m$ )
$\dot{m}$	Mass flow rate, ( $kg/s$ )
$Q_{conv}$	Heat convection rate, ( $Watt$ )
$q_s$	Heat Flux, ( $W/m^2$ )
$f$	Friction factor
$Nu$	Nusselt number
$Re$	Reynolds number
$Pr$	Prandtl number
$\Delta P$	Pressure difference
$T_b$	Bulk fluid temperature, ( $^{\circ}C$ )
$T_s$	Surface temperature, ( $^{\circ}C$ )
$T_w$	Wall temperature, ( $^{\circ}C$ )
$\Delta T$	Temperature difference
$\varepsilon$	Roughness size, ( $m$ )
$g$	Gravitational acceleration, ( $m/s^2$ )
$\phi$	Volume concentration of nanofluid
$A_s$	Surface area, ( $m^2$ )
$A_c$	Cross sectional area, ( $m^2$ )
$V_{avg}$	Average velocity, ( $m/s$ )
$V$	Voltage, (Volt)
$I$	Current, (Amphere)
$nf$	Nanofluid
$exp$	Experiment
$W$	Water

**LIST OF ABBREVIATIONS**

FYP	Final year project
htc	Heat Transfer Coefficient
Al	Aluminum
Cu	Copper
Ag	Silver
Au	Gold
Al <sub>2</sub> O <sub>3</sub>	Aluminum Oxide/Alumina
CuO	Copper Oxide
TiO <sub>2</sub>	Titanium Oxide
SiC	Silicon Carbide
DB	Dittus-Boelter

## **CHAPTER 1**

### **INTRODUCTION**

#### **1.1 PROJECT BACKGROUND**

The thermal conductivity of conventional fluids such as water is low. Nanofluids are engineered preparation of fluids with suitable surfactants to disperse metal particles of nanometer size in a liquid such as water. The use of nanofluids for possible commercial application as coolant in automobiles, electronic components is widely pursued. Experimental test rig has been designed and fabricated to evaluate forced convective heat transfer coefficient. The experimental values of Nusselt number obtained and will be compared with the values in literature.

#### **1.2 PROBLEM STATEMENT**

Nowadays, lots of industrial and consumer product have heat transfer as their most vital process. But there is a limit of the heat transfer due to the inherently poor thermal performance of conventional fluids and restricted in developing energy efficient heat transfer fluids that are required for ultrahigh performance cooling. The increases of the global competition force the need to build up superior heat transfer fluids with considerably higher thermal performance than are presently available. Hence, lots of research has been made by the scientists and engineers to break this fundamental limit by dispersing metallic and non-metallic nanoparticles in liquids.

Research in nanofluid technologies have offered a vast potential for further expansion of high performance, compact and cost effective cooling to utilizing in

industrial applications. Consequently, heat transfer behavior studies for nanofluids are vital in providing authentication result that have found by preceding researchers.

### **1.3 OBJECTIVES OF STUDY**

The objective of the study is as follow:

- i. To estimate the heat transfer coefficient (htc) of  $\text{TiO}_2$  water-based nanofluid flowing through a plain tube.
- ii. To investigate the effect of nanofluid concentration on forced convective heat transfer.

### **1.4 SCOPES OF THE STUDY**

There are several scopes included in this research as follow:

- i.  $\text{TiO}_2$  nanofluids dilute using distills water are flow under fully developed turbulent region.
- ii. Nanofluids flow through a plain tube at constant heat flux boundary conditions (402 W)
- iii.  $\text{TiO}_2$ /water nanofluids with volume concentration of 1 % to 3 vol. %
- iv. Evaluate htc under turbulent region with Reynolds number between 10,000 and 30,000.

### **1.5 SIGNIFICANCE OF STUDY**

Nanofluid is a fluid containing nanometer-sized particles, called nanoparticles. These fluids are engineered colloidal suspensions of nanoparticles in a base fluid. The nanoparticles used in nanofluids are typically made of metals, oxides, carbides, or carbon nanotubes. Common base fluids include water, ethylene glycol and oil.

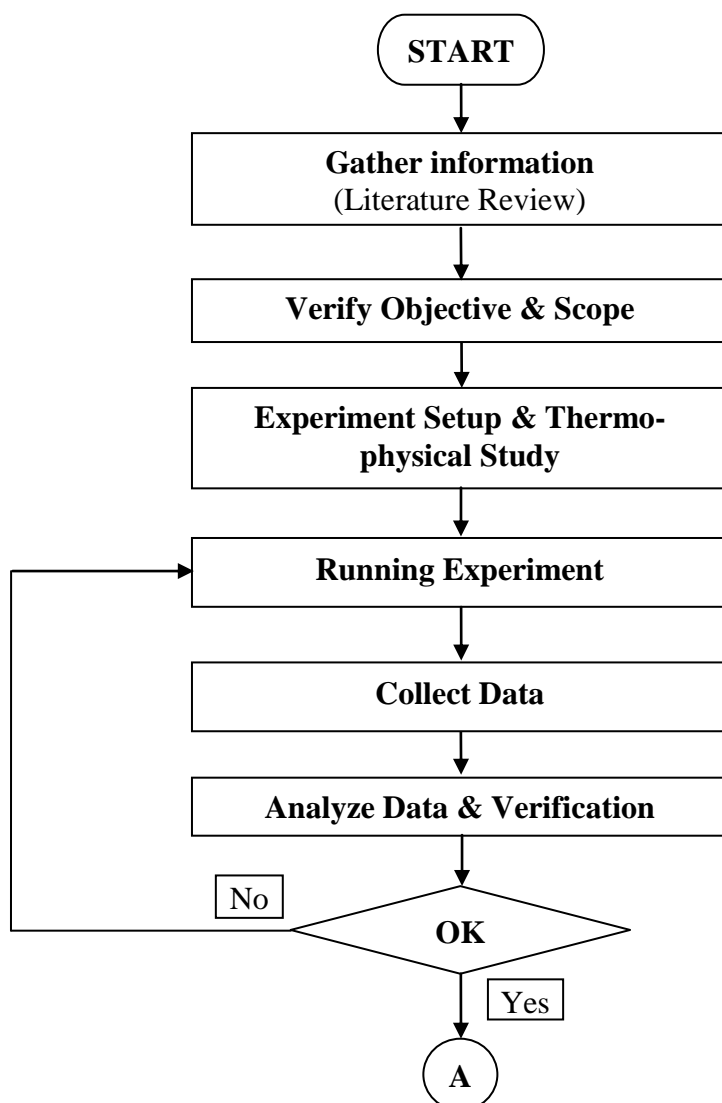
Nanofluids have novel properties that make them potentially useful in many applications in heat transfer, including microelectronics, fuel cells, pharmaceutical processes, and hybrid-powered engines, engine cooling/vehicle thermal management,

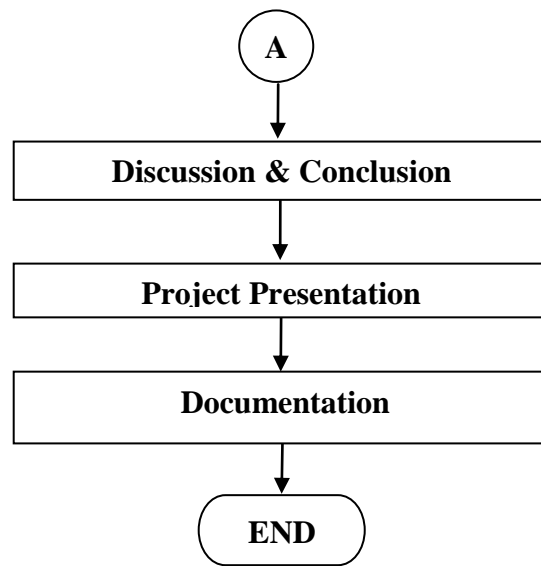
domestic refrigerator, chiller, heat exchanger, nuclear reactor coolant, in grinding, machining, in space technology, defense and ships, and in boiler flue gas temperature reduction. They exhibit enhanced thermal conductivity and the convective heat transfer coefficient compared to the base fluid.

Nanofluids thermal performance was highly affected by volume concentration, flow condition, constant heat flux and surface temperature of the wall tube. To produce nanofluids with an ideal composition, study of the force convection effect with varies parameter was essential.

## 1.6 PROJECT FLOW CHART

Figure 1.1 shows the process flow of the project with step by step start from beginning of the project until the end.





**Figure 1.1:** Project Flow Chart

## **CHAPTER 2**

### **LITERATURE REVIEW**

#### **2.1 BACKGROUND**

The enhancement of heat transfer using nanoparticles is explored through the work of researchers over the past 15-20 years. An introduction includes the definition of a nanofluid and the proposed approaches for analyzing such a system. A brief history of the work that's been done in the recent past and the conceptual reasoning for explaining experimental observations is also discussed. Current research is indicating the Forced Convective Heat Transfer Coefficient with Nanofluids in a Plain Tube. For this research there a need verified the equation to determine the viscosity, density, specific heat and thermal conductivity of the nanofluid. This is because the thermal properties of nanofluids are needed to find the Nusselt number of the nanofluids.

#### **2.2 VISCOSITY**

The turbulent friction and heat transfer behaviours of dispersed fluids with submicron  $\gamma\text{-Al}_2\text{O}_3$  and  $\text{TiO}_2$  particles were investigated experimentally by Pak and Cho, 1998 in a circular pipe. Viscosity measurements of those fluids were also conducted by using a Brookfield viscometer. The result of the study showed that the viscosity for the dispersed fluids with  $\gamma\text{-Al}_2\text{O}_3$  and  $\text{TiO}_2$  particles showed shear-thinning behaviour at or above volume concentration of 3% and 10%, respectively. At the 10% volume concentration, the relative viscosity for the dispersed fluid with  $\gamma\text{-Al}_2\text{O}_3$  particles were approximately 200, while it was approximately 3 for dispersed fluids with  $\text{TiO}_2$  particles. These viscosity results are significantly larger than the predictions from the classical theory of suspension rheology. The submicron size of



metallic particles in dispersed fluids plays a significant role in the determination of viscosity.

Namburu et al., 2007 discuss about the silicon dioxide nanofluids with ethylene glycol/water as base fluids exhibit non-Newtonian behaviour at lower temperatures. At higher fluid temperatures, the viscosity and shear rate relation does not change, proving Newtonian behaviour. The viscosity of SiO<sub>2</sub> nanofluids increases as the volumetric nanoparticle concentration increases. For example, the viscosity of 10% SiO<sub>2</sub> particle volume concentration is about 1.8 times the viscosity of the base fluid. As temperature increases, the viscosity of SiO<sub>2</sub> nanofluid decreases exponentially.

An experiment has been carried out by He et al., 2007 on the flow and heat transfer behaviour of aqueous TiO<sub>2</sub> nanofluids flowing through a straight vertical pipe under both the laminar and turbulent flow conditions. The effects of nanoparticles concentrations, particle (agglomerate) size, and the flow Reynolds number are investigated. Nanofluids used in this work show the shear thinning behaviour and the shear viscosity tends to be a constant at shear rates greater than 100 s<sup>-1</sup>. The constant viscosity increases with increasing particle concentration and particle size.

The effective viscosities of water-based nanofluids containing very low concentrations of Al<sub>2</sub>O<sub>3</sub> nanoparticles (Al<sub>2</sub>O<sub>3</sub> –water nanofluids) were experimentally investigated by Lee et al., 2008. The Al<sub>2</sub>O<sub>3</sub>–water nanofluids with various concentrations from 0.01 to 0.3 vol% using a two step method with ultrasonication and without any surfactant were used in this experiment. Viscosity measurements show that the viscosity of the Al<sub>2</sub>O<sub>3</sub> –water nanofluids significantly decreases with increasing temperature. The alumina nanofluids have a nonlinear relation between their viscosity and the nanoparticle concentration even at very low (0.01–0.3 vol%) concentrations, while the Einstein viscosity equation clearly predicts a linear relation.

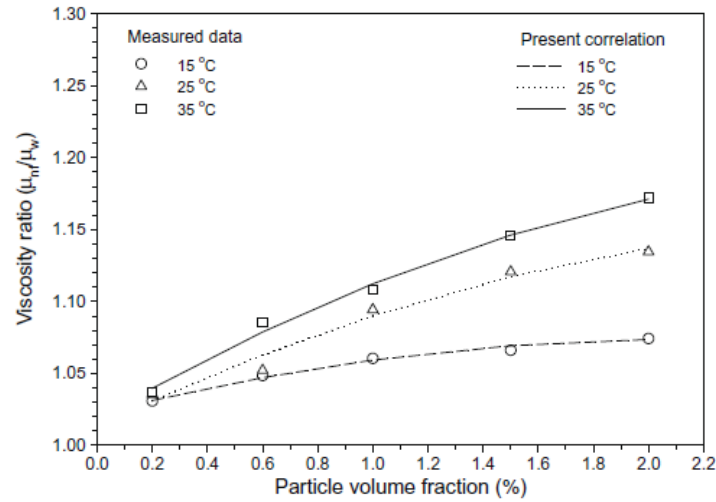
The viscosity of nanofluids was found to significantly increase with the particle volume fraction in Murshed et al., 2008. The viscosity was influenced by the particle size, interfacial layer, and volume fraction. The enhancement of viscosity may diminish the effectiveness of nanofluids in practical applications.

Nguyen et al., 2008 has established new and complete viscosity database for the water–Al<sub>2</sub>O<sub>3</sub> nanofluid with 36 and 47 nm particle sizes. The viscosity data have been obtained for the ambient condition and particle volume fraction varying from 1 to nearly 13%. The data were taken with varies temperature and particle-size, considering temperature ranging from 22 to 75 °C and particle volume fraction from 1 to 9.4%. It has been found that in general, the nanofluid viscosity strongly depends on both temperature and concentration; while the particle-size effect seems to be important only for sufficiently high particle fraction. The existence of a critical temperature has clearly been established. Beyond such a critical temperature, the particle suspension properties seem to be irreversibly altered, which, in turn, may induce hysteresis behaviour. The critical temperature has been found strongly dependent on both particle fraction and size. The hysteresis phenomenon observed on viscosity has raised serious concerns regarding the use of nanofluids for heat transfer enhancement purposes.

The effective viscosities of TiO<sub>2</sub>–water nanofluids with concentrations from 0.2 vol% to 3.0 vol% were measured by Turgut et al., 2009 at temperatures from 13 °C to 55°C. The results of the study showed that for low volume additions of nanoparticles for 0.2% vol. fraction, the viscosity values follow quite well the viscosity values of pure water with a decrease in viscosity with increasing temperature and may be predicted by the Einstein law of viscosity. But, for higher additions of TiO<sub>2</sub> nanoparticles, the Einstein law of viscosity failed to explain the large increase in viscosity values. At 13 °C the increase was as large as two times that predicted by the Einstein law of viscosity for 3% vol. fraction.

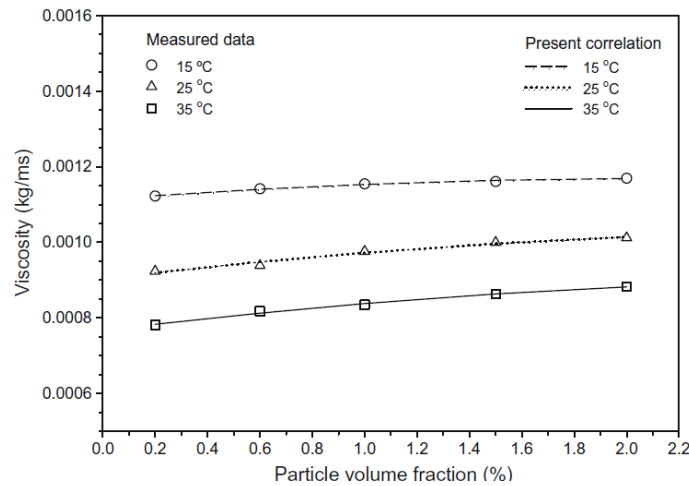
Duangthongsuk & Wongwises, 2009 investigated experimentally the effective thermal conductivity and effective viscosity of TiO<sub>2</sub>-water nanofluids with particle volume concentrations of 0.2, 0.6, 1.0, 1.5 and 2.0 vol%. The nanofluids viscosity was measured at the temperature ranging between 15°C and 35°C. The viscosity of nanofluids significantly decreases with increasing temperature and increases with increasing particle volume concentration. However, they states that the results of this study were different from the other researcher which may be caused by various parameters such as particle size, particle preparation (solution chemistry), the

measurement technique or even different particle sources. Finally, the proposed correlations for predicting viscosity of nanofluids show good agreement with the experimental results as shown in Figs. 2.1 and 2.2.



**Figure 2.1:** Comparison of the measured viscosity ratio with the proposed correlation.

Source: Duangthongsuk & Wongwises (2009)



**Figure 2.2:** Comparison of the measured viscosity with the proposed correlation.

Source: Duangthongsuk & Wongwises (2009)

The convective heat transfer performance and flow characteristic of TiO<sub>2</sub>-water nanofluid flowing in a horizontal double tube counter-flow heat exchanger was experimentally investigated by Duangthongsuk & Wongwises, 2010. Experiments were carried out under turbulent flow conditions. The effect of particle concentrations and the

Reynolds number on the heat transfer performance and flow behaviour of nanofluids are determined. The result of the experiment shows that the dispersion of the nanoparticles into the base liquid increases the thermal conductivity and viscosity of the nanofluids, and this augmentation increases with increasing particle concentrations. The pressure drop of nanofluids increases with increasing Reynolds number and there is a small increase with increasing particle volume concentrations. This is caused by increase in the viscosity of nanofluids, and it means that nanofluids incur little penalty in pressure drop.

In Kole & Dey, 2010 study, Alumina nanofluids based on a car engine coolant with excellent stability have been prepared and their thermal conductivity and viscosity both as a function of volume fraction of  $\text{Al}_2\text{O}_3$  nanoparticles and temperature between 10 and 80 °C have been investigated. The result showed that viscosity of the nanofluids increases with alumina volume fraction and decreases with the rise in temperature. The engine coolant with and without the surfactant is a Newtonian fluid. Newtonian behaviour is observed for nanofluids with low  $\text{Al}_2\text{O}_3$  loading ( $\ll 0.004$ ) only at higher temperatures, while nanofluids with higher  $\text{Al}_2\text{O}_3$  loading display non-Newtonian features throughout the measured temperature range. For low  $\text{Al}_2\text{O}_3$  loading ( $\ll 0.004$ ) at 30 °C, the nanofluid obeys the Bingham model, while for higher loading ( $> 0.004$ ) shear thinning characteristics are displayed. The analysis confirms the superiority of the expression derived by Masoumi *et al* over the classical models to predict the relative viscosity of an alumina nanofluid based on a car engine coolant.

### 2.3 THERMAL CONDUCTIVITY

Turbulent friction and heat transfer behaviours of dispersed fluids with submicron  $\gamma\text{-Al}_2\text{O}_3$  and  $\text{TiO}_2$  particles were investigated experimentally in a circular pipe. The Pak and Cho, 1998 study shows that the Nusselt number for the dispersed fluids increased with increasing volume concentration as well as Reynolds number. However, it was found that the convective heat transfer coefficient of the dispersed fluid was 12% smaller than that of pure water when compared under the condition of constant average velocity. Therefore, better selection of particles having higher thermal

conductivity and larger size is recommended in order to utilize dispersed fluids as a working medium to enhance heat transfer performance.

A research to find the effective thermal conductivities of fluids with  $\text{Al}_2\text{O}_3$  and  $\text{CuO}$  Nanoparticles dispersed in water, vacuum pump fluid, engine oil, and ethylene glycol was conducted by Wang et al., 1999. The research shows that the thermal conductivities of nanoparticle–fluid mixtures increase relative to those of the base fluids and a possible relation between the thermal conductivity increase and the particle size which is the thermal conductivity of nanoparticle–fluid mixtures increases with decreasing the particle size. The thermal conductivity increase also depends on the dispersion technique.

By using existing models for computing the effective thermal conductivity of a mixture, it is found that thermal conductivities computed by theoretical models are much lower than the measured data, indicating the deficiencies of the existing models in describing heat transfer at the nanometer scale in fluids. It appears that the thermal conductivity of nanoparticle fluid mixtures is dependent on the microscopic motion and the particle structure. Any new models of thermal conductivity of liquids suspended with nanometer-size particles should include the microscopic motion and structure-dependent behaviour that are closely related to the size and surface properties of the particles.

Lee et al., 1999 measured the thermal conductivity of four oxide nanofluids ( $\text{Al}_2\text{O}_3$  in water,  $\text{Al}_2\text{O}_3$  in ethylene glycol,  $\text{CuO}$  in water, and  $\text{CuO}$  in ethylene glycol) by a transient hot-wire method to experimentally investigate the thermal conductivity behaviour of dilute nanofluids. The results show that nanofluids, containing only a small amount of nanoparticles, have substantially higher thermal conductivities than the same liquids without nanoparticles. For the copper oxide/ethylene glycol system, thermal conductivity can be enhanced by more than 20 percent at a volume fraction of 0.04 (4 vol%). In the low-volume fraction range tested (up to 0.05), the thermal conductivity ratios increase almost linearly with volume fraction, but with different rates of increase for each system. The present experimental data also show that the thermal conductivity of nanofluids depends on the thermal conductivities of both the

base fluids and particles: For nanofluids using the same nanoparticles, the conductivity ratio increases of ethylene glycol nanofluid systems are always higher than those of water nanofluid systems; For nanofluids using the same liquid, the conductivity ratio of the CuO system is always higher than that of the  $\text{Al}_2\text{O}_3$  system.

Temperature effect of thermal conductivity enhancement in nanofluids by Das et al., 2003 shows an increase in thermal conductivity enhancement of nanofluids can take place over a temperature range of  $21^\circ\text{C}$  to  $51^\circ\text{C}$ . This finding makes nanofluids even more attractive as cooling fluid for devices with high energy density where the cooling fluid is likely to work at a temperature higher than the room temperature. It states that nanofluids containing smaller CuO particles show more enhancement of conductivity with temperature. However the enhancement is considerably increased for nanofluids with  $\text{Al}_2\text{O}_3$  as well. The study indicate that particle size is an important parameter for the observed behaviour and the usual weighted average type of model for effective thermal conductivity is a poor approximation of the actual enhancement particularly at the higher temperature range.

The effective thermal conductivity of nanofluids was found to significantly increase with the particle volume fraction. The proposed models by Murshed et al., 2008, which consider particle size, interfacial layer, and volume fraction, show good agreement with the experimental results and give better predictions for the thermal conductivity of nanofluids compared to the existing models. Besides the volume fraction of particle, it can also be concluded that particle size, shape, interfacial layer, and temperature also influence the thermal conductivity of nanofluids. The effect of temperature on the enhanced effective thermal conductivity of nanofluids is important for theoretical understanding and needs to be considered for the model development.

Lee et al., 2008 paper were experimentally investigated the thermal conductivities of water-based nanofluids containing very low concentrations of  $\text{Al}_2\text{O}_3$  nanoparticles ( $\text{Al}_2\text{O}_3$ -water nanofluids). The result of the experiment shows that the thermal conductivities of the dilute  $\text{Al}_2\text{O}_3$ -water nanofluids increase almost linearly with concentration. Furthermore, the measured thermal conductivities of the  $\text{Al}_2\text{O}_3$ -water nanofluids are consistent in their overall trend with previous data at higher

concentrations and agree well with the predicted values by the Jang and Choi model over a very wide concentration range from 0.01 to 5 vol%.

Beck et al., 2009 have measured the thermal conductivity of nanofluids containing seven sizes of alumina nanoparticles ranging from 8 to 282 nm in diameter. The results indicate that the thermal conductivity enhancement decreases as the particle size decreases below about 50 nm. The decrease in the thermal conductivity of the nanoparticles was because the particle size becomes small enough to be affected by increased phonon scattering. The limiting value of the enhancement for nanofluids containing large particles is greater than that predicted by the Maxwell equation, but is predicted well by the volume fraction weighted geometric mean of the bulk thermal conductivities of the two phases.

The thermal conductivity of TiO<sub>2</sub> nanoparticles in deionized water nanofluids was measured by Turgut et al., 2009 using a  $3\omega$  method for volume fractions ranging from 0.2% to 3.0%. The data showed that the thermal-conductivity enhancement was in relatively good agreement with the Hamilton–Crosser model. The  $3\omega$  measurement method for thermal conductivity was particularly well adapted as it required small amounts of sample size and was rapid and accurate (uncertainty within 2%). The measurements showed that there is no dependence related to temperature; the thermal conductivity increased by the same order of magnitude as the base fluid which is water.

The effective thermal conductivity of TiO<sub>2</sub>-water nanofluids with varies volume concentrations and temperatures were investigated experimentally by Duangthongsuk & Wongwises, 2009. The results show that the relative thermal conductivity of nanofluids increases with increasing particle volume concentration and slightly decreases with increasing temperature. It is also seen that the existing correlations for predicting the thermal conductivity of nanofluids gave lower values than the experimental values.

Kole & Dey, 2010 indicate that the addition of 0.035 volume fractions of Al<sub>2</sub>O<sub>3</sub> nanoparticles in the engine coolant enhances the thermal conductivity of the fluid. The enhancement in thermal conductivity of the nanofluid varies linearly with the volume fraction of the nanoparticles and reaches a maximum of 11.25% at 80 °C for the

nanofluid containing 0.035 volume fraction of  $\text{Al}_2\text{O}_3$  nanoparticles. The thermal conductivity expression derived by Prasher *et al* well predicts the observed nanoparticle volume fraction dependence of the thermal conductivity of the present nanofluid.

The convective heat transfer performance  $\text{TiO}_2$ -water nanofluid flowing in a horizontal double tube counter-flow heat exchanger was experimentally investigated by Duangthongsuk & Wongwises, 2010. The study shows that dispersion of the nanoparticles into the base liquid increases the thermal conductivity of the nanofluids, and this augmentation increases with increasing particle concentrations. At a particle volume concentration of less than 1.0 vol%, the use of  $\text{TiO}_2$ - water nanofluid gives significantly higher heat transfer coefficients than those of the base fluid. For example, at the 1.0 vol%, the heat transfer coefficient of nanofluids was approximately 26% greater than that of pure water. However, at the particle concentration of 2.0 vol%, it was found that the heat transfer coefficient of nanofluids was roughly 14% smaller than that of pure water for the given conditions.

## 2.4 SPECIFIC HEAT AND DENSITY

The variation of specific heat capacity with particle size is supported with the theoretical and experimental observations by Wang et al., 2006 who studied its effect on thermal conductivity. The values of bulk specific heat obtained with CuO nanoparticles from experiments are found to be in close agreement with the theoretical predictions, at temperatures lower than 225K for particles up to 50nm size. However for temperatures above 225K, the values from theory predicted a decrease for particle sizes upto 10nm. The specific heat capacity showed an increasing trend for particles below 10nm.

Wang et al., 2006 reasoned that for particles larger than 10nm the quantum effect can be neglected and the specific heat will decrease monotonically with particle size. For particles which are smaller than 10nm, the quantum effect will increase the specific heat capacity of the nanoparticles uniformly, and thus create an irregular behaviour for particles of different sizes. This observation is stated to be in conformity with the monotonic decreasing relation proposed by Prasher and Phelan, 1998 for particles larger than 10nm.